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## (54) BUTENE-1 POLYMER SHEETS

(71) We, PRINCETON CHEMICAL RESEARCH INC., a corporation organised and existing under the laws of the State of New Jersey, United States of America, of Post Office Box 652, Princeton, New Jersey 08540, United States of America, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The invention relates to butene-1 polymer compositions for use as a paper-like material and also to processes for the preparation of such materials.

The invention provides an opaque, paper-like material which comprises a sheet of (a) from 20 to 97% by weight of a butene-1 polymer having a molecular weight in excess of 20,000, an isotacticity as herein defined of at least 25% and a butene-1 content of at least 50% by weight and (b) from 3 to 80% by weight of a filler particles distributed in the butene-1 polymer, the surface of the sheet containing microprotrusions and exposed filler particles. The sheet may have a maximum tensile strength ratio of from 10:1 to 3:1 in mutually perpendicular directions.

The invention also provides a process for preparing an opaque paper-like material which comprises extruding a composition comprising (a) from 20 to 97% by weight of a butene-1 polymer having a molecular weight in excess of 20,000, an isotacticity as herein defined of at least 25% and a butene-1 content of at least 50% by weight and (b) from 3 to 80% by weight of a filler particles distributed in the butene-1 polymer, to form a sheet and stretching the sheet to at least 5% beyond its yield strain at a temperature of less than 60°C.

During the process the sheet is opacified and rendered ink-retentive. Advantageously, the sheet is stretched in the machine and transverse directions at least 20% beyond its

yield strain. The filler should be normally solid and insoluble in the polymer.

Suitably pigmented or dyed compositions will develop colour contrasts in comparison to the unstretched compositions. Due to the stretching a large alteration in the surface properties of the material results so that in addition to the characteristics of the polymer base, the surface now also exhibits characteristics and physical properties of the filler or fillers. In other words, the sheet now presents a heterogeneous surface. For example, when a sheet of polybutene-1 filled with about 50% of calcium carbonate is suitably stretched according to the method of the present invention, the surface becomes receptive and retentive to conventional printing or writing inks.

The butene-1 polymers useful for the invention are produced preferably by a Ziegler-type polymerization of butene-1 alone or butene-1 with olefinically unsaturated monomers to provide polymers having molecular weights of about 20,000 to 2,000,000. As comonomers ethylene, propylene, styrene, butadiene, isoprene and  $C_6-C_{10}$  alpha-olefins may be used but the homopolymer of butene-1 is preferred.

The butene-1 polymers should preferably have an isotacticity of greater than 50%. The isotacticity as herein defined is measured by the percentage by weight of the polymer which remains insoluble in diethyl ester. The filled polybutene-1 composition should preferably contain from 25 to 90% of butene-1 polymer.

The type and amount of filler can be changed to obtain specific properties in the stretched polybutene material. Thus, a fine particle size of less than 10 microns, e.g. calcium carbonate having a 2 micron average size or less, provides a smooth-textured white opaque material with excellent ink-retentivity useful for a bond paper substitute; a large particle size calcium carbonate, for example,

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7 microns or more in average particle size, provides a rough-textured white opaque material with excellent ink-retentivity. Also, a fibrous filler such as wollastonite provides a rough-textured white opaque material with excellent ink-retentivity, a tissue-like appearance, and a paper-like "crackle". The use of reinforcing fillers such as ultrafine or precipitated silicas, carbon blacks, high-styrene styrene-butadiene copolymers, and the like, give increased stiffness, or "crackle". The fillers can also be used to change or enhance other properties of the stretched and opacified sheets, such as colour, degree of flexibility, gloss, smoothness, strength, tear resistance, flame retardancy or barrier properties. The fillers may be inorganic or organic solids, and can be selected so as to modify the properties and obtain specific surface characteristics of the films or sheets.

Inorganic fillers include common natural and synthetic fillers and pigments, such as kaolin, bentonite, and other clays; carbonates and silicates of calcium, magnesium and barium; oxides and silicates of aluminium, zinc, titanium, lead and zirconium, hydroxides of magnesium and aluminium; mica, talc, pumice, asbestos, and wollastonite; barium and calcium sulphates; different types of silica such as amorphous, diatomaceous, crystalline, reinforcing, ultrafine and pyrogenic silica; carbon blacks; hydrated aluminium silicates; sodium aluminium fluoride; powdered, microspheroidal and fibrous glass; graphite; iron oxide; lithopone; and pyrophyllite. For special purposes powdered or metal flakes such as aluminium iron, or zinc may also be used.

The organic fillers include solid polymers such as polystyrene or styrene copolymers; vinyl chloride homopolymers and copolymers; polyethylene, including ultra-high molecular weight polyethylenes; polypropylene; polymethyl-1-pentene; ethylene copolymers and terpolymers; polyacrylic and polymethacrylic acid and esters including copolymers; cellulose esters, microcrystalline cellulose and colloidal cellulose; synthetic rubbers based on butadiene and styrene and based on ethylene and propylene; nitrile polymers and terpolymers such as acrylonitrile-butadiene-styrene; wood flour, cotton linters; polyamides, polyethers and polyurethanes.

In addition to the butene-1 polymer and the inorganic and organic fillers, there may be added to the composition one or more plastic and rubber compounding additives. These additives include anti-oxidants or pro-oxidants, thermal stabilizers, ultraviolet stabilizers, optical brighteners or depressants, waxes, hydrocarbon resins, metal stearates, tackifiers and lubricants. Further, pigments and dyes can be incorporated and foaming agents can also be employed for specific effects.

When polymeric organic fillers are used, the polymeric organic filler portion of the composition should not exceed 150 parts per hundred parts by weight of butene-1 polymer, and it is preferred that the polymer filler portion be less than 110 parts per hundred parts by weight of butene-1 polymer. When used, the level of polymeric filler is preferably at least 5 parts, more preferably, at least 10 parts per 100 parts of butene-1 polymer by weight.

The inorganic filler portion of the composition should not exceed 80 weight percent of the total filled polybutene composition and it is preferred that the total composition contain no more than 70 weight percent of inorganic filler. The minimum level of inorganic filler can be 3% and is preferably 5%, when no other fillers are used.

The compositions of this invention can be prepared using such equipment as two-roll mills, Banbury mixers, twin-screw extruders, or shell blenders. It may be practical to mix and blend the polymeric materials together in a first step when polymeric fillers are employed, and then to incorporate any inorganic materials and the other organic additives into the blended polymeric composition. Alternately, the entire blending operation can be performed in a single step. The composition may be formed into a sheet or film by flat die extrusion, blow extrusion, or film casting followed, if desired, by calendering. For special applications, sheets may be also fabricated by compression or injection moulding.

Although films or sheets of filled butene-1 polymer compositions can be oriented at temperatures from about 90°C up to close to the crystalline melting point of the polymer, stretching the filled butene-1 polymer compositions of the present invention above 90°C does not yield the paper-like properties which are characteristic of the present novel articles. The method of the present invention requires that the stretching be conducted at a temperature of less than 60°C; preferably as low as -25°C, more preferably between 10°C and 40°C, and most preferably between 20°C and 35°C. Stretching is conveniently performed as the last stage in the fabrication process and, although monoaxial stretching yields useful products, it is preferred to stretch the sheets or films biaxially. For example, extruded film may be cooled on a chill roll and stretched by passing through "stretch" rolls operating at higher speed than the chill roll. Tentering frames may be used and stretching may also be effected by means of gas pressure. As has been noted, stretching modifies the transparency and surface characteristics of the filled butene-1 polymer compositions. The extent of stretching should be sufficient to cause the desired degree of opacification and modification of surface properties. It should be recog-

nized that the incorporation of fillers particularly at high loadings such as more than about 50%, results in reducing both the yield strain and the elongation at break. The yield strain refers to the extent of stretching beyond which the sheet will no longer return to its original dimension when released. The greatest alteration of surface properties occurs when the compositions are stretched to an elongation just below their breaking point, e.g. about 75 to 90% of their breaking elongations. The compositions may be stretched immediately after fabrication into sheets or films, or they may be aged for periods up to one year and more before stretching. The rate of stretching is preferred to be the highest rate which is obtainable and controllable with the available equipment; for example, in practice, films can be stretched at rates up to 5,000% elongation per minute, but rates as low as 10% per minute can also yield useful products. When stretched monaxially, the compositions generally exhibit a higher tensile strength in the direction of stretching, i.e. the machine direction, than in the transverse direction. With biaxial stretching, the ratio of tensile strengths can be made to approach the value of unity. For products which are to be used as paper substitutes, it is usually desired that the maximum ratio of tensile strengths measured in mutually perpendicular directions within the plane of stretched sheet or film be less than 3:1, and for many uses, as close to 1:1 as is practical. However, products of the present invention are useful for many applications when this maximum tensile strength ratio is as high as 10:1.

After stretching, the paper-like sheets may be further processed into substitute textile articles or other products. For example, the sheets may be adhesive-bonded, stapled, sewn or heat-sealed. In the case of heat-sealing, e.g. along an edge of a paper substitute sheet, the sealed region will suffer a loss in paper-like properties, but the major part of the surface of the article, which has not been exposed to the heat-sealing temperature, retains its opacity and printability.

The butene-1 polymer compositions of the present invention have enhanced tensile strength, water and moisture resistance, wet strength, tear resistance, creep resistance, grease resistance, and other excellent barrier properties, which are inherent properties of the base resin.

When exposed to outdoor solar radiation, products of the present invention can be made to degrade by the use of suitable stabilizer systems and thermal treatments. Thus, the products of the present invention can be made disposable by outdoor degradation without harm to or pollution of the environment. Products of the present invention are useful as substitute writing and printing papers. Such

uses include the printing of consumer magazines; trade and technical journals; reference books and directories; mail order catalogues; direct mail and trade catalogues; books, particularly children's books which require superior tear resistance, factory manuals and books used outdoors which require enhanced water-, grease-, or oil-resistance; book dust jackets; greeting cards; documents such as driver's licences and passports; labels and tags, especially for outdoor use; magazine and catalogue covers; menus; cooking recipe cards; printed advertising, handbills, billboards, posters, signs, and notices; maps, wall charts, and wall paper; and art reproduction prints. Other uses include graphic, music, drafting, and art paper; adding machine, duplicating, ledger, envelope, diazo, manifold, onion skin, xerographic, lithograph and offset paper computer paper; photographic print paper; filing cards; data retrieval cards, file folders, and data processing cards; bond paper, letter-writing paper, typewriting paper, carbon paper, air mail paper, personal stationery, exercise books, school tablets; banknote, check and stamp paper. Other uses of the paper-like products of the present invention are in packaging. These applications include grocery, shopping and packaging bags; refuse and garbage bags; tissue wrap for flowers and shoes; inserts for hosiery and apparel; handbag stuffing; food wrap such as for individual fruit, meats, fish and particularly greasy foods such as butter, margarine, sausage, cold cuts, baked goods, cheeses, and carry-out hamburgers and french fries; glassine paper substitutes; pouches for dry soup and sauce mixes; cereal box liners and soda cracker inner wraps; vegetable parchment; frozen food wrap; cigarette packs; record sleeves; shipping wrap; heavy duty bags such as for cement, fertilizer, feedstuff chemicals, etc.

Other embodiments of the present invention are useful as substitute textile articles, such as napkins, wiping cloths and table cloths; hospital and stewardess' gowns, physician's gloves; bibs and aprons; curtains; disposable clothing such as swimsuits, baby pants and diaper liners; rainwear and protective clothing, seatcovers and headrest covers and non-woven fabrics.

The paper-like sheets of the invention will be particularly described with reference to the drawings wherein:

Figure 1 is a scanning electron micrograph of the surface of a sheet before stretching not according to the invention, the micrograph having been taken at a 45° angle of incidence of the electron beam at 10,000× magnification prior to photographic enlargement;

Figure 2 is a similar view of the sheet of Figure 1 after stretching which is according to the invention;

Figures 3 and 4 are similar views of another specimen, before and after stretching, at a magnification of only 500 $\times$ ; and

Figure 5 and 6 are views corresponding respectively to Figures 3 and 4 but at 10,000 $\times$  magnification.

The sheet of Figures 1 and 2 contained 47.4% polybutene-1, 47.4% calcium carbonate, 5% polystyrene, and 0.2% stearic acid (all percentages are by weight). The micrographs were taken at 10,000 $\times$  magnification before photographic enlargement, using a 45° angle of incidence of the electron beam. Stretching has created micropores in the surface of the sheet, the average diameters of the pores (dark regions on the micrographs) ranging from about 0.5 to 0.01 microns or less. The individual calcium carbonate filler particles are also evident. Before stretching, Figure 1, the filler particles are apparently substantially covered by a membrane consisting of the polybutene-1 resin phase, while after stretching Figure 2, the membrane has ruptured around many of the individual particles in the surface, exposing the particles. Thus the unstretched surface is essentially homogeneous, while the stretched surface contains both micropores and exposed filler particles. The combination of micropores and exposed filler particles are believed to account for the excellent ink-receptivity and ink-retentivity of the products of the present invention, and also contribute to the enhanced opacity. Figure 3 involves a composition somewhat richer in filler, the view lacking some of the detail of Figure 1 because of the lower magnification. Figure 4 shows almost no micropores, they being barely visible because of their small size. When enlarged 20 $\times$  further to the scale of Figures 1 and 2, the micropores are readily visible in Figure 6 which is the stretched product shown in Figure 5 before stretching.

The invention is illustrated by the following Examples in which all parts are by weight.

#### Example 1.

Polybutene-1 (47.4 parts) having a melt index of 0.8 and 43 percent diethyl ether insolubility was mixed with 47.4 parts of calcium carbonate having an average particle diameter of 2 microns and range of 0.3 to 8 microns; 5 parts polystyrene, and 0.2 parts of stearic acid on a two-roll mill at 107°C. The milled sheet was cut into small pieces and fed to a Dreher grinder to obtain pellets. The polybutene pellets were extruded in a one-inch Killion extruder, at an extrusion temperature of about 200°C, through a rod die to obtain rod of about  $\frac{1}{2}$  inch diameter. The rod extrudate was cooled by pulling through a water-filled trough by a chill roll. The rod was cut into 12-inch lengths and was fed again to the Dreher grinder to obtain

pellets. The polybutene pellets were extruded in a one-inch Killion extruder, at a temperature of about 210°C, through a 6 inch film die. The film extrudate was dropped onto a water-cooled chill roll to obtain 15 mil film. The grey translucent film made opaque about 30 minutes after extrusion by holding one end of the film in a vice and pulling the other end by means of callender rolls at an extension rate of 600% per minute to obtain 4  $\times$  24-inch lengths of white opaque film. The stretching operation was conducted at room temperature. The laboratory experiment number and the date of the experiment were written on the white opaque and also on unstretched material using a ball point pen. The ink could not be erased from the stretched white opaque film by rubbing with a finger. The ink was erased readily from unstretched film of the same composition by rubbing over the letters with a finger. The product is shown in Figures 1 and 2, before and after stretching, respectively.

#### Example 2.

46.9 parts of polybutene-1, having a melt index of 0.8 and 93% ether insolubles was blended with 46.9 parts of calcium carbonate, 5 parts of polystyrene, 0.2 parts of stearic acid, and 1 part of ultrazinc yellow (derived from tetrachloro-isoindolone), using a two-roll mill at 108°C. The milled sheet was cut into small pieces, and fed to a Dreher grinder to obtain pellets. The pellets were rodlike as in Example 1, the rods ground to pellets, and then extruded through a 6-inch film die to give film 18 mils thick. A piece was cut from the film and compression-moulded at 170°C into a sheet 5 mils thick. The sheet was then cut into strips 1-inch wide. A strip was then placed in an Instron (Registered Trade Mark) tensile testing machine with an initial jaw separation of 1 inch. The strip was then stretched at the rate of 20 inches per minute to an elongation of 150%. This corresponds to a stretching rate of 2000% per minute. The elongation at break was about 170%.

Before stretching, the sheet was a glossy, translucent, yellowish-brown colour, and was not retentive to ink or pencil. After stretching, the sheet was non-glossy, pale yellow, and opaque, and was receptive and retentive to ink and pencil writing.

A second strip was similarly stretched at a rate of 20% per minute, to an elongation of 125%. After stretching, the sheet was similarly plate yellow and opaque, and was receptive and retentive to ink and pencil writing.

#### Example 3.

Example 1 was repeated using calcium carbonate having an average particle size of 7 microns and a particle range of 0.3 to 40 microns. The composition was converted into

a rough-textured white opaque film with excellent retentivity to pencil, ink and type-writing.

#### Example 4.

- 5 Example 1 was repeated using 34.9 parts of polybutene having a melt index of 1.8, 59.9 parts of calcium carbonate having an average particle size of 2 microns, 5 parts of polystyrene and 0.2 parts of stearic acid.
- 10 The composition was converted into a white opaque film with excellent ink retentivity.

- Figures 3 and 4 are scanning electron micrographs of the surface of the composition before and after stretching, respectively, taken at 500 $\times$  magnification before photographic enlargement, and at 45 $^\circ$  incidence for the electron beam. Figures 5 and 6 are corresponding electron micrographs of the sheet before and after stretching, respectively, taken at 10,000 $\times$  magnification before photographic enlargement. This formation of micropores and the exposure of individual calcium carbonate particles in the stretched composition are clearly evident.

#### Example 5.

- 25 Example 1 was repeated using 74.8 parts of polybutene-1 having a melt index of 1.8, 20 parts of wollastonite, 5 parts of polystyrene and 0.2 parts of stearic acid. The composition was converted into a rough-textured white opaque film with excellent ink retentivity.

#### Example 6.

- 35 Example 1 was repeated using 74.8 parts of polybutene-1, 20 parts of ultrafine precipitated silica, 5 parts of polystyrene and 0.2 parts of stearic acid. The composition was converted into a stiff paper-like white opaque film.

#### Example 7.

- 40 Example 1 was repeated using 59.9 parts polybutene-1, 34.9 parts of calcium carbonate, 5 parts of polystyrene, and 0.2 parts of stearic acid. The filled polybutene composition was converted into a smooth-textured white opaque film having excellent ink retentivity.

This paper substitute is useful as a type-writing and drawing paper.

#### Example 8.

- 50 Example 1 was repeated using 66.5 parts of polybutene-1, 33.3 parts of calcium carbonate and 0.2 parts of stearic acid. The filled polybutene composition was converted into a

white opaque film having excellent ink retentivity and opacity. This product is useful as a book printing paper. The tensile strength of the sheet in the stretched direction was 11,000 psi.

#### Example 9.

Polybutene (50 parts) having an isotactic content of 93% and a melt index of 2.3 was masticated in a Brabender Plastic-Corder at about 150 $^\circ$ C with 25 parts of calcium carbonate and 25 parts of high density polyethylene. The materials were masticated to obtain a homogeneous product. The masticated blend was moulded at 160 $^\circ$ C in a Pasadena press to a translucent white sheet having a thickness of 10 mils. The sheet was stretched manually the same day, beyond its yield point, to a white opaque sheet of 5 mils. The white opaque sheet had excellent ink retentivity. Before stretching, the sheet was not ink retentive. This composition can be used for freezer wrap.

#### Example 10.

Example 9 was repeated with 53 parts of butene-1 homopolymer, 23.5 parts of polyvinyl chloride and 23.5 parts of calcium carbonate. With the sides clamped to hold the sheet to constant width, the moulded sheet of the composition was stretched manually the same day at room temperature from 4 $\frac{1}{8}$  inches long and 11.2 mils thick to 5 $\frac{1}{8}$  inches long and 9 mils thick. The laboratory experiment and the date of the experiment were written on the stretched white opaque sheet and on the unstretched translucent sheet. The ink was erased from the unstretched sheet but could not be erased from the stretched white opaque sheet.

#### Examples 11, 12 and 13.

The composition given in Table I were blended according to the method given in Example 9, and then molded at 160 $^\circ$ C to give sheets having a thickness of 10 mils. None of these compositions contained inorganic fillers.

Strips of the moulded sheets were stretched either manually or by means of an Instron tensile testing machine. Examples 11 and 12 were identical, except that the strips were aged at room temperature for less than 1 day, and for three months, respectively. In all cases the samples were stretched at room temperature beyond their yield points.

As shown in Table I, Examples 11, 12 and 13 were ink-retentive after stretching.

TABLE I

Examples 11 to 13

Parts by weight

Example No.	11	12	13
Polybutene-1,2,3 melt index	69.3	69.3	65
Polyvinyl chloride	30.7	30.7	—
Polymethacrylonitrile	—	—	15
Ethylene-propylene copolymer	—	—	20
Ageing period between moulding and stretching	less than 1 day	3 months	less than 1 day
Ink-retentivity after stretching	retentive	retentive	retentive
Transparency before stretching	transparent	transparent	translucent
Transparency after stretching	opaque	opaque	opaque

## Example 14.

5 79.8 parts of polybutene-1 resin with a melt index of 1.8 was blended with 15 parts of pigment-grade talc, 5 parts of polystyrene, and 0.2 parts of stearic acid, on a two-roll mill at 107°C. The composition was extruded into film, as in Example 1, 3 mils thick. The film was stretched at room temperature at an elongation rate of 800% per minute to give an opaque white sheet.

10 The paper-like sheet is useful as food wrap and shopping wrap.

## Example 15.

15 50 parts of polybutene-1 having an isotactic content of 93% and a melt index of 2.3 is masticated in a Brabender Plasti-Corder with 25 parts polypropylene of inherent viscosity 1.64 and crystallinity 97%, and 25 parts of calcium carbonate, at about 165°C to obtain a homogeneous product. The blend is moulded at 160°C to form a white translucent sheet 4 mils thick.

20 The sheet is stretched to an elongation of 140% in one direction, and then to an elongation of 110% in the perpendicular direction. The resulting paper-like sheet is opaque and ink retentive and is useful as a food wrap.

## WHAT WE CLAIM IS:—

1. An opaque, paperlike material which comprises a sheet of (a) from 20 to 97% by weight of a butene-1 polymer having a molecular weight in excess of 20,000, an isotacticity as herein defined of at least 25% and a butene-1 content of at least 50% by weight and (b) from 3 to 80% by weight of a filler particles distributed in the butene-1 polymer, the surface of the sheet containing micropores and exposed filler particles.

2. A material according to claim 1 in which the sheet has a maximum tensile strength ratio of from 10:1 to 3:1 in mutually perpendicular directions.

3. An opaque, paper-like material substantially as shown and as described with reference to Figures 2, 4 or 6.

4. An opaque, paper-like material substantially as described with reference to any one of the Examples.

5. A process for preparing an opaque paperlike material which comprises extruding a composition comprising (a) from 20 to 97% by weight of a butene-1 polymer having a molecular weight in excess of 20,000, an isotacticity as herein defined of at least 25% and a butene-1 content of at least 50% by

- weight and (b) from 3 to 80% by weight of a filler particles distributed in the butene-1 polymer to form a sheet and stretching the sheet to at least 5% beyond its yield strain at a temperature of less than 60°C. 15
- 5 6. A process according to claim 5 in which the filler particles in the sheet have a particle size of less than 10 microns and form from 5 to 80% by weight of the composition.
- 10 7. A process according to claim 5 or claim 6 in which the filler particles comprise an organic solid.
8. A process according to any of claims 5 to 7 in which the filler particles comprise an organic polymer and form at least 10% by weight of the composition. 15
9. A process according to claim 5 or claim 7 in which the filler particles comprise an inorganic solid.
10. A process according to any of claims 5 to 9 in which the sheet is stretched in two transverse directions to at least 20% beyond its yield strain. 20

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COMPLETE SPECIFICATION

3 SHEETS

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Sheet 1

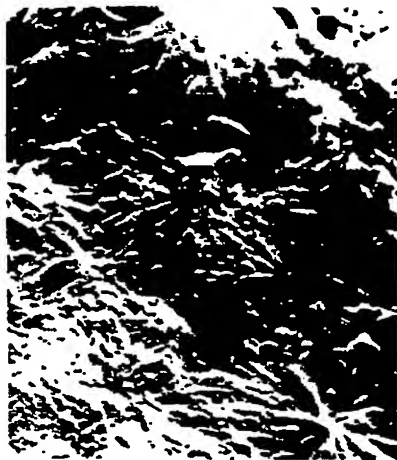


FIG. 1

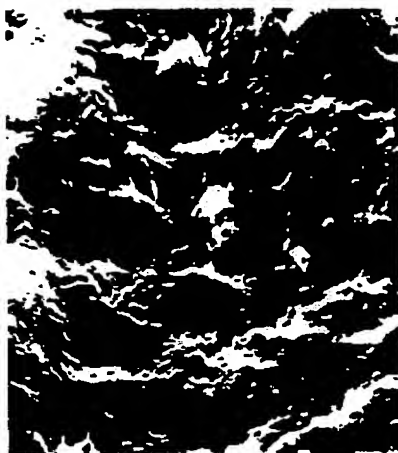


FIG. 2





FIG.3



FIG.4



FIG. 5

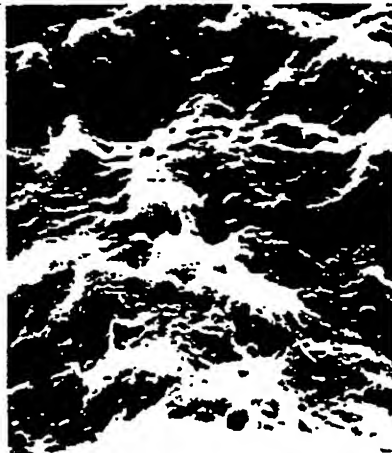


FIG. 6

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